

Response to Reviewers for ar-2025-40 “Soot growth by monodisperse particle dynamics model coupled with Computational Fluid Dynamics” by Fakharneshad, Berry and Goudeli. The Reviewers’ comments are shown in *italics*. The responses (blue) and changes in the revised manuscript (red), are given below. Corrections made on this new revision were highlighted in red.

Reviewer 1

The manuscript compares molecular-dynamics-derived nucleation, coagulation, and surface growth rates with empirical rate models in a premixed ethylene burner-stabilized stagnation (BSS) flame, focusing on their influences on the modeled particle number concentration and volume fraction. This is accomplished by solving the coupled mass, energy, and momentum transport equations using Fluent. Overall, this study is potentially interesting and relevant to the Aerosol Research. However, the presentation would benefit from significant improvement to better highlight the value of the work. The following comments are provided for the authors’ consideration and focus on selected aspects, although the authors are encouraged to carefully revise the manuscript as a whole, beyond the specific points listed below.

1. The methodology section would benefit from a more structured organization, for example by clearly separating geometry, governing equations, boundary conditions, and initial conditions. In addition, the descriptions should be more complete. For instance, although the temperature conditions can be inferred from Figure 1, all initial and boundary conditions for the flow, energy, and transport equations should be explicitly stated in the text. The manuscript provides the transport equations. It would also be beneficial to provide the exact flow and energy equations, as they are critical.

1. Thank you for this suggestion. The geometry, governing equations, and boundary conditions are separated in the methodology section, and the governing equations section has been modified in the revised manuscript:

“2.1.2 Governing equations

The steady-state Navier-Stokes equations were solved using a pressure-based solver that ensures mass conservation through a pressure correction equation. The equation for conservation of momentum (Batchelor, 2000) is:

$$\frac{\partial}{\partial t}(\rho \vec{v}) + \nabla \cdot (\rho \vec{v} \vec{v}) = -\nabla p + \nabla \cdot (\bar{\tau}) + \rho \vec{g} + \vec{F} \quad (1)$$

where ρ is the gas density, \vec{v} is the velocity vector, p is the static pressure, and $\bar{\tau}$ is the viscous stress tensor. The terms $\rho \vec{g}$ and \vec{F} represent the gravitational and external body forces, respectively. The energy equation was also included to capture thermal variations:

$$\frac{\partial}{\partial t} \left(\rho \left(e + \frac{v^2}{2} \right) \right) + \nabla \cdot \left(\rho v \left(h + \frac{V^2}{2} \right) \right) = \nabla \cdot \left(k_{eff} \nabla T - \sum_j h_j \vec{J}_j + \bar{\tau}_{eff} \cdot \vec{v} \right) + S_h \quad (2)$$

The e and h on the left-hand side of eq. 2 denote the internal energy and the enthalpy, respectively. On the right-hand side, k_{eff} is the effective thermal conductivity, \vec{J}_j is the diffusion flux of species j , and S_h accounts for volumetric heat sources.”.

A new “Initial conditions” section has also been added in the revised manuscript:

“2.1.4 Initial conditions

All simulations were first initialized using the Hybrid Initialization method used to generate a physically representative initial flow field through boundary-based interpolation. Then, Fluent simulations were performed at cold-flow conditions (no combustion chemistry considered) until the flow reaches steady state. Upon convergence of the cold-flow solution, the volumetric reactions were activated, and a temperature of 2000 K was imposed in the entire region above the burner surface. This temperature patch acts as an ignition source to initiate flame formation in the combustion zone.

This two-step procedure ensured numerical stability and provided a physically realistic initial condition for the reacting flow simulations.”

2. *The manuscript would benefit from a clearer description of how the MD-derived rates are obtained. While readers may refer to the cited literature for details, it would be helpful to briefly summarize the underlying chemical and physical mechanisms, the types of systems used to derive these rates, and the conditions under which the rates are applicable.*

2. Thank you for this suggestion. The following sentences have been added in the revised manuscript: “... were described using MD-derived expressions. The expression for the MD-derived nucleation rate (Model II) was developed from MD simulations of isothermal acetylene pyrolysis conducted at a flame-relevant temperature range of 1200–1800 K and high-pressure of 189–568 atm (Fakharneshad et al., 2025). The nucleation rate was determined by monitoring the temporal evolution of incipient soot cluster formation relating it to the initial acetylene concentration (Table 1: $S_{N, Nucl.}$ of Model II).”

3. *The capabilities and limitations of the monodispersed model should be clarified. As currently described, it appears that the model is able to represent only the total particle number concentration, rather than size-resolved particle distributions. It would be beneficial to have more structured and involved discussions on this.*

3. Indeed, the particle size distribution is not resolved as the present model assumes a monodisperse population of particles/agglomerates/aggregates at each time (or height above the burner), as described in the fourth paragraph of the Introduction of the original manuscript: “The particle dynamics equation can be greatly simplified ... for coupling with multidimensional CFD simulations in realistic geometries.” This simplification increases the computational performance of the PD model when implemented in CFD as equations are solved only for one size class. However, the self-preserving mobility size distribution can be generated based on the average mobility diameter (Fig. 7) if the self-preserving geometric standard deviation is known.

To further address the Reviewer’s comment, the following discussion has been added in revised manuscript: “... compared with the average d_m derived from particle size distributions measured by the scanning mobility particle sizer (SMPS) in premixed ethylene flames (Camacho et al., 2015). Even though the present PD model does not account for the agglomerate/aggregate polydispersity, the mobility-based self-preserving size distribution can be derived based on the average d_m (Fig. 7) and the mobility-based self-preserving geometric standard deviation of soot ($\sigma_{g,m} = 1.48 \pm 0.03$), obtained by discrete element method simulations (Kelesidis and Goudeli, 2021: Fig. 2).”

4. *In Line 105, Section 3.1.1, the text shows “Error! Reference source not found.” Please provide the correct reference.*

4. It has been fixed.

5. *In Line 124, the parameter C is introduced without a clear definition. In Equation (3) as well. Please clarify if C represents the total concentration of carbon species, or is individual concentration for different carbons.*

5. We thank the reviewer for pointing this out. Here, C represents total molar concentration of carbon contained in soot particles (obtained by converting the soot mass concentration using the molar mass of carbon, $MW_c = 12$ g/mol), rather than the concentration of individual gas-phase carbon species. To clarify this, we have modified 1st paragraph of Section 2.2 of the revised manuscript: "... the total carbon molar concentration C of carbon contained in soot particles (obtained by the soot mass concentration using the molar mass of carbon, $MW_c = 12$ g/mol) was modelled using CFD ...".

6. In Equations (2) and (3), the notation used for the source terms on the right-hand side may be misleading, as they resemble derivatives of the variables N and C , although they are not (they are source terms). Please consider revising the notation to avoid potential confusion.

6. The notation for source terms in Equations (2) and (3) has been changed in the revised manuscript: "...the transport of total particle number density N is described by:

$$\frac{\partial(\rho u_i N)}{\partial x_i} - \frac{\partial}{\partial x_i} \left(\Gamma_{k,N} \frac{\partial N}{\partial x_i} \right) = S_{N, Nucl.} + S_{N, Coag.} \quad (4)$$

and the transport of carbon molar concentration C is defined by:

$$\frac{\partial(\rho u_i C)}{\partial x_i} - \frac{\partial}{\partial x_i} \left(\Gamma_{k,C} \frac{\partial C}{\partial x_i} \right) = S_{C, Nucl.} + S_{C, SG} \quad (5)$$

The terms on the left-hand side of equations (4) and (5) represent convection and diffusion in the flow, where u_i denotes the velocity, and $\Gamma_{k,N}$ and $\Gamma_{k,C}$ are the effective diffusion coefficients for number density and carbon concentration, respectively. The right-hand side of eq. (4) includes the nucleation term, $S_{N, Nucl.}$, which describes the formation of new particles, and the coagulation term, $S_{N, Coag.}$, which describes the reduction in the number of particles due to agglomeration. In eq. (5), the source terms correspond to carbon addition due to newly formed particles by nucleation, $S_{C, Nucl.}$, which can be obtained by:

$$S_{C, Nucl.} = S_{N, Nucl.} \times \frac{n^*}{N_{Av}} \quad (6)$$

where n^* is the number of carbon atoms within the soot critical nucleus and N_{Av} is Avogadro's number. $S_{C, SG}$ is the accumulation of carbon on existing particles due to surface growth.".

7. In Line 140, the variables Wc and M should be defined when they are introduced.

7. Thank you. The MW_c is one variable corresponding molar mass of carbon. To avoid confusion, the clause has been modified in the revised manuscript: "... volume of a primary soot particle, $V = \frac{C \times MW_c}{\rho_{soot}}$ is the total soot volume concentration of the whole particle population and $MW_c = 12$ g/mol...".

8. Several variables listed in Table 1 have not been defined elsewhere in the manuscript. Please ensure that all variables appearing in the table are clearly defined.

8. A Nomenclature section has been added in the revised manuscript.